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# PATENT

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

Confirmation No. 9412

Appl. No. : 10/716,894  
 Applicant(s) : Cornelis F. VAN EGMOND *et al.*  
 Filed : November 19, 2003  
 TC/A.U. : 1764  
 Examiner : In Suk C. BULLOCK  
 Title : Methanol and ethanol production for an oxygenate to olefin reaction system  
 Atty. Docket No. : 2003B113  
 Customer No. : 23455

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

DECLARATION SUBMITTED UNDER 37 C.F.R. § 1.132

**Dear Sir:**

I, Teng Xu, a U.S. citizen, residing at 13422 Scenic Glade Drive, Houston, TX 77059,  
declare and state that:

1. I received a Ph.D. in Chemistry at the University of Texas A&M University.
2. Since 1998, I have engaged in scientific research at ExxonMobil Chemical Company in the field of Methanol Conversion to Olefins on Molecular Sieve Catalysts.
3. I am author or co-author of several publications dealing with converting oxygenates such as methanol to olefins on zeolite solid acids.
4. I am one of the joint inventors of U.S. Patent Application No. 10/716,894 (the '894 application) and am familiar with the subject matter thereof.

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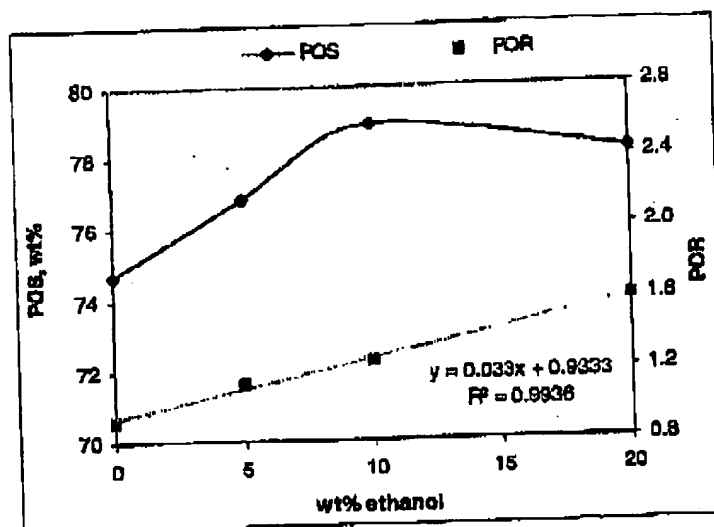
5. I have read U.S. Patent No. 6,441,262 (Fung) cited in the July 17, 2006 Office Action and am familiar with the subject matter thereof.

6. The Fung reference discloses using a silicoaluminophosphate molecular sieve catalyst to convert a combination of methanol and ethanol to light olefins. The reference is particularly concerned with the problem of how to moderate the olefin product content in an alcohol conversion reaction in which the alcohol feed is predominantly methanol. Fung addresses this problem by using two contact zones. One zone is referred to as an oxygenate conversion zone, which is where a combination of unregenerated and regenerated catalyst contacts methanol for conversion to olefin. The other zone is referred to as an alcohol contact zone, which is where ethanol, propanol or butanol is contacted with regenerated and fresh catalyst. The use of this dual contacting zone allows for manipulating the olefin content in the product stream.

7. This invention is particularly concerned with maximizing the amount of light or prime olefin formed (i.e., ethylene plus propylene), and at the same time further maximizing the amount of ethylene in the product. This problem is solved by providing a mix of methanol and ethanol, at a 4:1 to 19:1 weight ratio of methanol to ethanol, to a reaction zone, and contacting that mix with a silicoaluminophosphate molecular sieve catalyst at a temperature of at least 475°C to 500°C, and possibly higher, in the same reaction zone.

8. Aside from the data in Examples II and III (especially Tables III and IV) of the '894 application, additional experiments were performed under my authority and control over the claimed range (4:1 to 19:1 weight ratio of methanol to ethanol, particularly at 5 wt% ethanol to augment the data already described for 10 wt% and 20 wt% ethanol), following the procedures of Examples II and III. The experiments were performed at 475°C, 100 WHSV, and 25 psig (total pressure of mixed alcohol feed), the same density for methanol and ethanol being assumed in calculating WHSV. The results are shown in the following graph:

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10. From the data in the Examples of the '894 application and the data shown in the above graph, I conclude that:
- the amount of ethanol in methanol feed has a significant impact on POR (prime olefin ratio, i.e., ethylene to propylene ratio);
  - there is almost a linear relationship between wt% ethanol in feed vs. POR;
  - the amount of ethanol in methanol feed has a minor impact on POS (prime olefin selectivity, i.e., percent of ethylene + propylene in product); and
  - POS shows a peak beginning at about 5 wt% ethanol and extending at least up to about 20 wt% ethanol.
11. I further conclude from the data that at a temperature of at least 475°C to 500°C, a mix of from 5 wt% to 20 wt% ethanol provides superior POS and POR, and that such results would not have been expected by reading the Fung reference.
12. It is also my opinion that the coupling of temperature with the particular mix of ethanol and methanol as a feed stream would to provide superior POS and POR is not mere optimization of a reaction process, because there are enough significant variables and the

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complexity of the reaction process is so high that one would not have been able to easily reach such a finding.

13. I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the captioned application or any patent issued therefrom.

Further declarant sayeth not.

11/20/2006  
Date

Teng Xu  
Teng Xu